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Review

Nutrient Water Pollution from Unsustainable Patterns of Agricultural Systems, Effects and Measures of Integrated Farming

Roxana Maria Madjar [1], Gina Vasile Scăețeanu [1,*] and Mirela Alina Sandu [2]

[1] Faculty of Agriculture, University of Agronomic Sciences and Veterinary Medicine of Bucharest, 59 Marasti Blvd., District 1, 011464 Bucharest, Romania
[2] Faculty of Land Reclamation and Environmental Engineering, University of Agronomic Sciences and Veterinary Medicine of Bucharest, 59 Marasti Blvd., District 1, 011464 Bucharest, Romania
* Correspondence: gina.scaeteanu@agro-bucuresti.ro

Abstract: Nowadays, agricultural practices require special attention due to their important contribution to water pollution, the more so as they are associated with environmental and health impairments. Despite legislation addressing nutrient pollution, there are still high levels of nutrients in water bodies, as evidenced by the results identified in the literature. Among nutrients of environmental concern identified in water and associated with agricultural practices are nitrogen and phosphorus. When applied in excess under fertilizer form, these nutrients accumulate in water bodies with consequences such as eutrophication or human health impairments if water is used for drinking. The purpose of this review paper is to present the current state of nutrient water pollution generated by unsustainable agricultural practices. In addition, are presented the main legislative regulations addressing water quality imposed by the European Union, are described aspects related to nitrogen and phosphorus pollution from agriculture, and levels of nutrients in water bodies from different European countries. Also, effects of agricultural pollution on the environment and humans are discussed, and possible strategies that could be employed to decrease or prevent water pollution are reviewed.

Keywords: agriculture; ammonium; eutrophication; farming; fertilizers; nitrate; nitrite; phosphorus; regulations; water framework directive

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1. Introduction

One of the sustainable development goals (SDG) set by the United Nations is SDG 6, which is based on ensuring availability and sustainable management of water and sanitation for all. In 2024, the SDG progress report highlighted that none of the SDG 6 targets are on track to be met. Nevertheless, between 2015 and 2022 it was recorded an increase from 69% to 73% of population that use safely managed drinking water, but global progress on implementing integrated water resources is still slow: 49% in 2017, 57% in 2023, very far from the target to be reached in 2030 (91%) [1].

Deterioration of water bodies is determined by population-accelerated growth, climate change (high temperatures, droughts), and intemperate use of inputs in agriculture. The use of fertilizers and pesticides is a great determinant of high yields, and their application must be managed carefully, but even though there are negative impacts on the environment, and on water sources as well. For instance, fertilizers contain, among other elements, nitrogen and phosphorus necessary to plant growth, but excessive application or not at the right time results in accumulation in surface and groundwater bodies. Furthermore, most common agricultural inputs (nutrients, pesticides, manures from livestock farms, etc.) are the most prevalent nonpoint sources of water pollution [2].

This review paper presents a systematization of literature data regarding water pollution resulted from agricultural sources, dealing with nutrients (nitrogen and phosphorus), legislation addressing nutrient pollution, the effects on the environment, and the potential measures that may influence decreasing water pollution from agricultural practices.

In order to achieve the objectives of our paper, an analysis of the specialized literature was conducted to identify the most relevant works that complete the review's plan with information. Consequently, databases were used, such as Clarivate Analytics' Web of Science (WoS), Elsevier's Scopus (Scopus), and Science Direct, for searching papers. The following keywords and/or combinations of keywords have been used: "water nutrient pollution", "water pollution from agriculture", "nitrogen" + "phosphorus" + "water pollution", "agricultural inputs"," eutrophication". This procedure helped to identify several categories of papers (reviews, reports, and communications), from which the suitable ones have been selected for this review and classified using the paper's chapters and subchapters as criteria. Furthermore, the reference lists of these papers were scanned to collect further related papers. As a result, the initial database was enriched by the snowballing technique with additional papers, which were used for extracting data.

Resulted information was further included in the review paper, emphasizing the pollution of water with nutrients (nitrogen and phosphorus) resulted from agricultural practices.

2. Nutrient Pollution

Agricultural pollution with nutrients is a global threat, and reducing it is challenging because the application of inputs is beneficial for crops and farmers. Among nutrients of primary environmental concern related to water pollution are nitrogen and phosphorus, and their presence in soil could be problematic when fertilizer application exceeds plant needs.

Global consumption of nitrogen fertilizers has skyrocketed, increasing from 18.66 million metric tons in 1965 to a staggering 109.17 million tons in 2021. Concerning phosphorus fertilizers, it was reported also an increasing trend of consumption, but more moderate than for nitrogen, starting from 16.02 million metric tons in 1965 to 47.81 million metric tons in 2021 (Figure 1) [3].

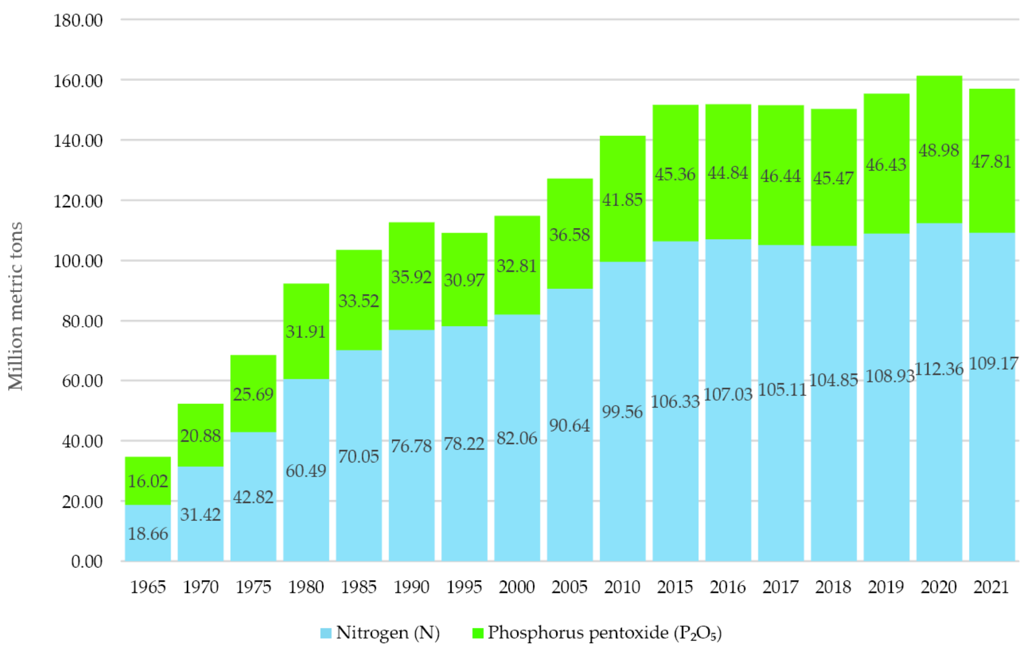

Figure 1. Global consumption of agricultural fertilizer from 1965 to 2021, by nutrient (in million metric tons).

It is therefore important that soil nutrients are properly managed because, when applied excessively, they either remain in the soil or enter aquatic environments, affecting water quality. Proper management of nitrogen and phosphorus is important from both agronomic and environmental perspectives, the more so as, once released to the environment, these nutrients are costly to control.

According to recent studies [4,5], nitrogen and phosphorus are already exceeding safe boundaries, with researchers being involved in finding measures and solutions to reduce nitrogen and phosphorus pollution.

2.1. Legislation Aspects Addressing Nutrient Levels in Water

The water framework directive (2000/60/EC) (WFD) [6] establishes a framework for protection of inland waters, transitional waters, coastal waters, and groundwater. Hence, it contributes to the prevention and decreasing of water pollution, the main objective being the achievement of good environmental status for all waters.

WFD is composed of more targeted directives [7], as it is systematized in Figure 2.

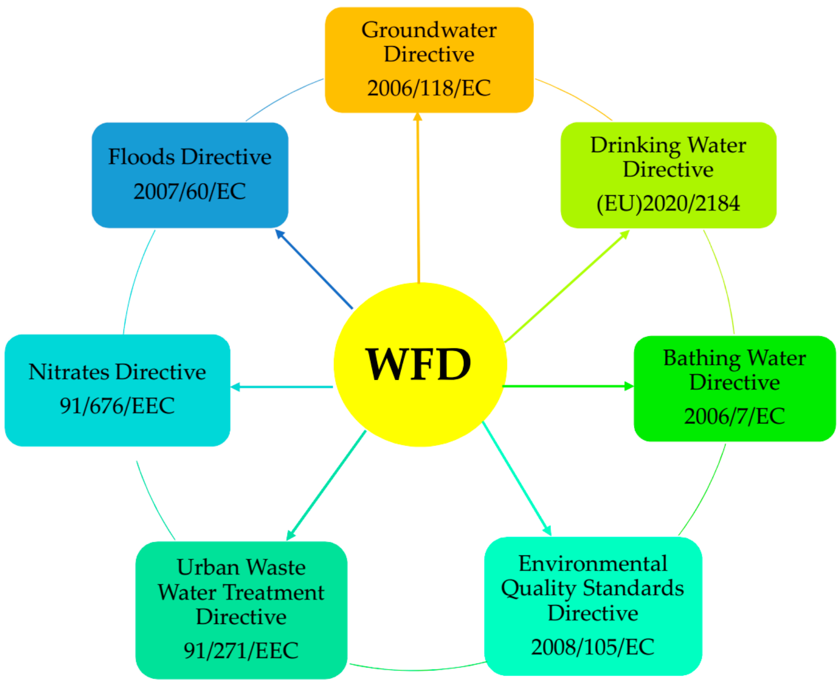

Figure 2. Water Framework Directive and supporting water directives.

Nitrates from agricultural practices may pollute drinking water sources and groundwater, and in the case of surface water, the main consequence is eutrophication. In 1991, the European Union initiated its nitrates directive (91/676/EEC) [8], which aims to protect waters from nitrates resulting from agriculture. According to Nitrates Directive [8], EU member states must designate nitrate vulnerable zones (NVZs). These are areas that contaminate with nitrates in surface water or groundwater with at least 50 mg L^{-1}. In addition, in these areas, there is a limit of 170 kg ha^{-1} year $^{-1}$ of nitrogen resulted from organic manure applied by farmers.

The presence of nitrogen species is drinking water in regulated by groundwater directive [9] and drinking water directive (DWD) [10], and the maximum admitted levels are 0.5 mg·L^{-1} for ammonium (NH_4^+) and nitrite (NO_2^-) and 50 mg·L^{-1} for nitrate (NO_3^-). In addition, member states shall ensure that the relationship [nitrate]/50 + [nitrite]/3 $\leq$ 1, where the square brackets signify the concentrations (mg·L^{-1}) for nitrate and nitrite, is complied with and that the parametric value of 0.10 mg·L^{-1} for nitrites is respected with ex water treatment works. In addition, EU Directive 2006/118/EC [9] suggests that member states should set quality standards, develop methodologies for assessment and monitoring of groundwater quality, and implement measures supporting groundwater protection.

WFD [6] was transposed into law in EU member states, and on the basis of biological and physico-chemical monitoring results was associated with the ecological status (high, good, moderate, poor, and bad). Furthermore, WFD indicates that EU member states should get involved to achieve good ecological status for all surface water and groundwater bodies by 2027. The original target for implementing the WFD terms was in 2015, or at least 2021.

In addition, the classification of surface water quality must consider the recommendations based on the Working Group ECOSTAT report [11]. This report provides a comparison of the nitrogen and phosphorus limits for lakes and rivers used by EU Member States for implementation of WFD. According to this, nutrients' concentrations should be determined in relation to individual types of lakes and rivers and must provide a good ecological status for freshwaters (rivers and lakes) under WFD. Limit values provided by EU states vary between countries, regions, and even parts of the catchments.

According to Romanian legislation [12], the quality of surface water is evaluated on the basis of nutrients' levels, and according to their concentrations' range five quality classes were established (Figure 3).

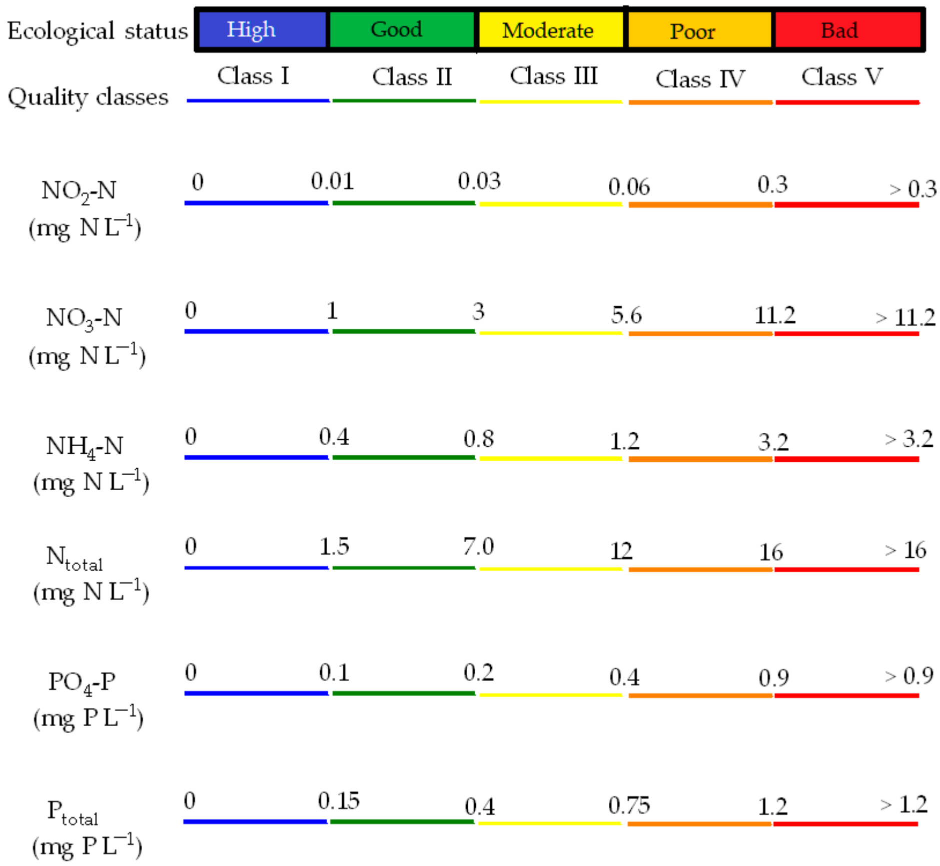

Figure 3. Ecological status and quality classes according to nutrients' levels for the classification of surface water quality.

2.2. Nitrogen Pollution

2.2.1. Nitrogen Forms and Route from Agriculture to Water Bodies

Nitrogen (N) is a macronutrient with great importance in growth and development of all living tissue, in crop plants and yield formation, being involved in plant metabolism, photosynthesis, and its application increases the greenness of the plants, the quality of crop yields, and the CO_2 assimilation rate [13].

Nitrogen is encountered in soils as organic compounds and in inorganic species as ammonium (NH_4^+) and nitrate (NO_3^-) ions, with different processes occurring between them (Figure 4).

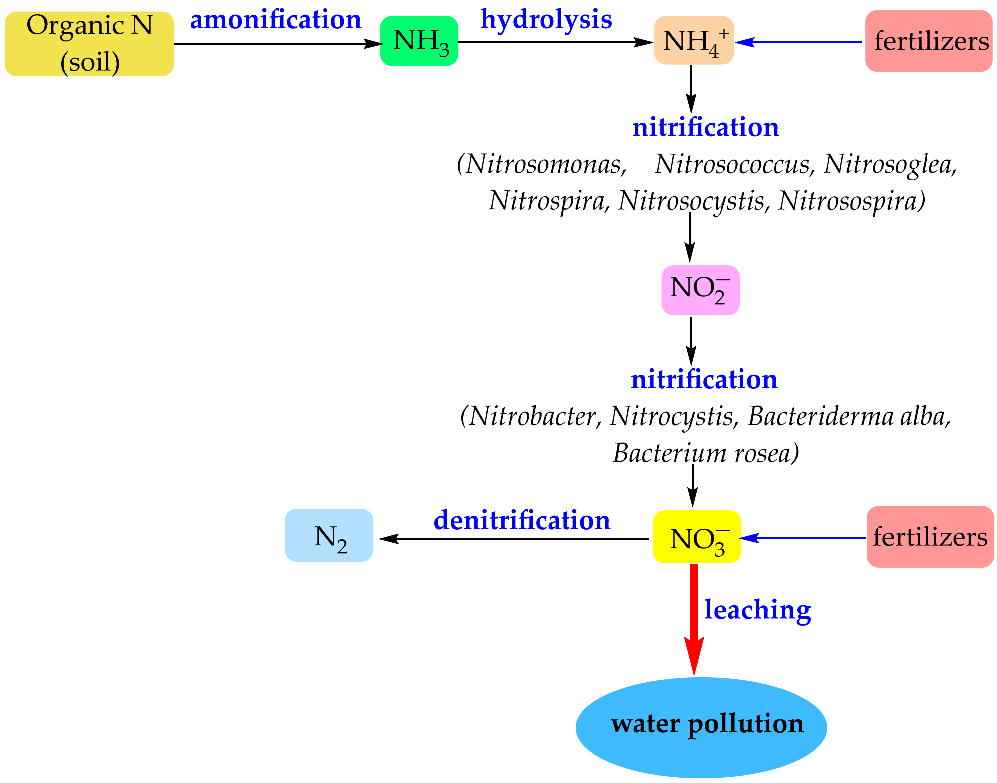

Figure 4. Nitrogen transformations in soil and water pollution.

Organic compounds with nitrogen under bacterial activity are decomposed in polypeptides, peptides, and amides, finally resulting in ammonia. In certain conditions, ammonia is converted by nitrification into nitrites and nitrates, according to the chemical reactions depicted below [14]:

(a) $2NH_3 + 3O_2 \rightarrow 2HNO_2 + 2H_2O + 168$ kcal
(b) $HNO_2 + ½O_2 \rightarrow HNO_3 + 42$ kcal

Nitrate ions being negatively charged will not adhere to negatively charged soil particles and consequently leach easily. Ammonium instead, even if it is positively charged and retained easily by the soil adsorption complex, is converted by oxidation into nitrate via nitrite.

When nitrogen inputs applied in the soil exceed plants' needs, excessive nitrates enter either groundwater or surface water.

The continuously increase in nitrogen fertilizer usage has led to groundwater and surface water pollution with nitrogen-containing species, mainly nitrates. From applied nitrogen fertilizer, about 50% is used by crops, the other part is mineralized and afterwards used by plants, and the rest is lost by leaching [15]. Even though leaching is the main way through which nitrates accumulate in water, soil erosion is also reported [16].

According to different studies, almost 80% of nitrogen species present in aquatic environments stem from agriculture [17].

2.2.2. Discrimination of Nitrogen Sources in Water

Stable isotope analysis is used by researchers to obtain data regarding the origin, transport, and transformation of nitrogen species in water.

For identification and prediction of water enrichment with nitrogen species resulted from different sources, variation of natural nitrogen ($\delta^{15}N$) and oxygen ($\delta^{18}O$) isotopes is used. Based on $\delta^{15}N$ values, it is possible to distinguish between natural and synthetic fertilizers, which are characterized by different isotopic signatures. For example, $\delta^{15}N$-NO_3 ranges from −8 to 7‰ for inorganic fertilizers, from +6 to +30‰ for organic fertilizers, and from +5 to 35‰ for manure, while for soil organic matter ranges from +3 to +8‰. In the case of ammonium fertilizers, $\delta^{15}N$-NH_4 signatures are reported to be 2.5‰ lower than signatures of $\delta^{15}N$-NO_3 from synthetic fertilizers [18].

The combined information provided by the dual isotopic approach ($\delta^{15}N$ and $\delta^{18}O$) may reveal the sources of nitrate from water, even though there are many limitations [19]. Consequently, Li and co-workers [20] have identified the variability of nitrate in the case of Changjiang River on the basis of $\delta^{15}N$ and $\delta^{18}O$ values, the nitrate sources being precipitation, chemical fertilizers, nitrification of organic materials, and sewage sludge.

In another study [21], a comparison of the isotopic composition of ammonium ($\delta^{15}N$-NH_4) and nitrate ($\delta^{15}N$-NO_3) in the Yeongsan River combined with hydrological modeling was used to track water contamination sources. Also, on the basis of $\delta^{15}N$ and $\delta^{18}O$ isotopes, mineral fertilizers were found as main pollution sources for the Yellow River basin, the Songhua River basin from China, Lake Winnipeg from Canada, or the Guadalhorce River basin from Spain [15].

On the basis of $\delta^{15}N$ and $\delta^{18}O$ isotopes, it was possible to discriminate among nitrogen sources in Uinta Mountains lakes where high nitrate levels were detected. As fertilizers were not applied in that area, their presence is attributed to atmospheric transport from agricultural regions [22].

Furthermore, on the basis of isotopic data, ammonium or urea-based nitrogen fertilizers were predominant sources of nitrate (30–61%) in the case of shallow soil layers from apple orchards. This study was undertaken to understand the pattern of nitrate leaching and prediction of nitrate pollution [23].

Although the use of $\delta^{15}N$ and $\delta^{18}O$ isotopes represents a useful tool to discriminate between different sources of pollution, lately the isotopic signature of boron ($\delta^{11}B$) was added to them and provided additional information. This approach is based on the fact that nitrogen and boron co-migrate in the groundwater, the latter being unaffected by nitrification/denitrification processes that occur to nitrogen [24]. For instance, a study [25] developed during 18 years based on discrimination between pollution sources for the Seine River confirmed that boron isotopes traced anthropogenic inputs in the environment.

Also, simulation models are used to understand pollution pattern, predict them, and adopt proper strategies for preventing undesirable effects of pollution [26–28]. Evaluation of nitrate leaching from different soil types in Slovenia under different management scenarios was done on the basis of the SWAT model. The model was validated on the basis of soil moisture, evapotranspiration, fertilization, and nitrate leaching. It has been found that vegetable rotation causes on all investigated soils the most nitrate leaching [29].

2.2.3. Nitrogen Levels Resulted from Agricultural Practices in Water Bodies from Europe

Considering that about 50% of the applied nitrogenous fertilizers drain from the fields and contaminate surface and groundwater [30], many literature studies have been carried out during time with emphasis on water pollution by agricultural practices. Hence, scientists from European countries reported in specialized literature the levels of nitrogen species in groundwater and surface water (ponds, lakes, and rivers), and the results of some studies and reports are presented below.

- *Groundwater*

In the case of well waters collected in different agricultural areas from Romania, nitrate levels varied a lot, but the reported values were high. For example, water samples collected from Chiajna, Letca Nouă, and Mănăstirea detected nitrate concentrations as average 66.75, 92.66, and 120.77 mg L^{-1}, respectively [31]. Also, for Brănești, Săhăteni, and Matca, the detected concentrations were even tenth times higher than the limit of 50 mg L^{-1} set by the drinking water directive (DWD) [10], this being a consequence of intensive use of fertilizers, as authors indicated [32]. High values of groundwater nitrate, slightly above 150 mg L^{-1}, were identified in Covasna [33]. In Sălaj [34], nitrate levels were 334.07 mg L^{-1}, this being also a consequence of fertilizer abuse.

Considering nitrite levels, values determined for well water from Covasna, Romania [33] were 0.210 and 0.309 mg L^{-1}, which are below the limit of 0.5 mg L^{-1}, set by the drinking water directive (DWD) [10]. Also, for well water from the Muntenia region of Romania, the reported average values are 0.295 mg L^{-1} (Chiajna), 0.241 mg L^{-1} (Letca Nouă), and 0.125 mg L^{-1} (Mănăstirea) [31].

The groundwater quality in Spain is also alarming, since around 23% of groundwater bodies have nitrates higher than the risk threshold level established as 75% of the parametric value of 50 mg L^{-1}). Considering high nitrate values, Spain has developed a national strategy to overcome this situation. On the basis of the PATRICAL model, it has been deduced that 90% of groundwater bodies with high nitrate levels will be recovered in the next 6–12 years if the total applied nitrogen will decrease by 30% and nitrogen losses will be reduced to 50% [35].

During 2000 and 2019, agricultural impacted springs in the German Land, Saarland, and Rhineland-Palatinate were monitored for quantification of nitrate levels. The nitrate levels in 2000 varied between 20 and 40 mg L^{-1}, but since EU WFD entered into force, it has been observed that nitrate discharges from agricultural land have not decreased. Consequently, nitrate levels in water have increased during the monitoring period [36].

Nitrate levels from a set of 36 wells from the Azores archipelago, Portugal, ranged between 0.02 and 37.4 mg L^{-1}, with the highest values being associated with agricultural practices [37].

Evaluation of nitrate levels for well water from 3 different locations in Northwest Croatia pointed out the influence of agriculture on nitrate levels. For example, in location Koprivnica, an area characterized by intensive agricultural activities, the nitrate level was 28.7 mg L^{-1}, followed by Krizevci wells with 26.5 mg L^{-1}. In the case of location Kalnik without agricultural influence, the nitrate concentration was 4.6 mg L^{-1} [38].

The status of the public water supply from the Pleven administrative region of Bulgaria, characterized by modern intensive agriculture (grains and technical crops), was reported by Bankova [39]. During 2010–2017, in eight settlements nitrate concentrations were more than 50 mg L^{-1}, whereas water supplied for three settlements contained, on average, more than 100 mg L^{-1}.

A study that aims to monitor a ten-year trend (2008–2017) of nitrate levels in water in the public water systems and private wells from Mačva district of Serbia was conducted by Srećković and her team [40]. Nitrate values in public water supplies ranged (as average) between 1.6 and 41.9 mg L^{-1}, the highest determined value being 73 mg L^{-1}. Considering nitrate detected in private wells, average values ranged between 9.9 and 61.3 mg L^{-1}.

The assessment of nitrate levels from drinking water originating from groundwater in the Prefecture of Thessaloniki, Greece, evidenced values that ranged between 1.4 and 141 mg L^{-1}. From total analyzed samples, 7.7% exceeded value of 50 mg L^{-1}, whereas for 19.2% of samples the values were between 25 and 50 mg L^{-1}. This situation was due to nitrogen fertilizers that were used for agricultural purposes [41].

Furthermore, evaluation of nitrogen-containing species in drinking water from Greece as a first assessment of the implementation of Directive 98/83/EC [42], which was the former version of Directive (EU) 2020/2184 [10], was done by Karavoltsos and his team [43]. Nitrate levels above 50 mg L^{-1} were found in Northern Greece, which is characterized by intensive agriculture. The highest nitrite levels were found also in Northern Greece, but determined values were below 0.5 mg L^{-1}. Considering ammonium levels, the highest concentrations were found in samples collected from Cyclades islands (1.2 mg L^{-1}, as average; the highest value was 5.7 mg L^{-1}), followed by Peloponnese and the Dodecanese islands. The exceeding values were related to water contamination by sewage and fertilizers.

Later, in 2015, the Court of Justice of the EU ruled that Greece violated EU law concerning protection of water against pollution generated by nitrates resulted from agriculture and imposed a fine of 2,639 euros per day on Greece for each day since the initial April 2015 ruling that Greece has failed to comply [44].

The impact of agriculture on nitrate levels in groundwater has been pointed out for Austria by Cepuder and Shukla [45]. According to their study, 34% of monitored sites from Tullnerfeld contain over 70 mg L^{-1} nitrate, even though the use of nitrogen fertilizers decreased after 1970.

Intensive farming in the Mediterranean region of Sicily, Italy, has strongly impacted the quality of groundwater. From a number of 38 wells included in the study developed by Pisciotta and co-workers [46], over 20 exceeded the guideline value of 50 mg L^{-1}, the highest concentration being about 400 mg L^{-1}.

Monitoring of groundwater quality in Denmark is very important since more than 62% of the total land area is used for agriculture. For example, studies carried out during 1998 and 2004 evidenced that 16.9% of wells contained nitrate over the maximum allowed level and about 60% had nitrates lower than 1 mg L^{-1}. Since 1979, Danish farmers have adopted different agricultural measures, which led to a nitrate-declining trend [47].

Nitrate levels in groundwater from Malta are high, and this is mainly because half of the country's land area is taken up by intensive agriculture. The determined nitrate nitrogen concentrations were even almost 100 mg L^{-1}. Considering nitrite nitrogen, the highest concentration was slightly over 0.5 mg L^{-1}, whereas ammonium nitrogen was <0.02–0.211 mg L^{-1} [48]. Contrariwise, nitrate concentrations in groundwater from Finland are lower than in other European countries, and in 2012–2015 the threshold of 50 mg L^{-1} nitrate was exceeded in four agricultural and forestry areas [49].

Concluding, even though there are various variables that affect the nutrient levels (seasonal variability in flow rate, periods of drought, and rain quality), it is obvious that agricultural practices influence negatively the quality of groundwater. According to the data presented above, the levels of nitrate are in many cases over 50 mg L^{-1}. It is indisputable that the first measure to decrease nitrogen levels in groundwater is related to the limitation of nitrogen fertilizer use, but the effects are not immediate.

- *Surface water*

In a long-term study developed over a period of 24 years, 13 different sites from the Ebro River, Spain were monitored. The results of the survey indicated 4.4–19.0 mg L^{-1} nitrate nitrogen and 0.1–0.6 mg L^{-1} ammonium nitrogen [50]. High levels of nitrate expose the surface water to eutrophication, while ammonium and nitrogen endanger fish life, the more so as water pH is higher [51].

Considering nitrite–nitrogen level in Timis River, Romania, the values determined by Dunca [52] ranged (as average) between 0.01 and 0.171 mg L^{-1}, whereas values detected for Bega River, Romania, were lower (0.007–0.056 mg L^{-1}). In addition, nitrite nitrogen levels in water collected from Moara Domnească pond, Romania, were as high as water was framed in Vth quality class (0.322 mg L^{-1}). Furthermore, ammonium levels were also very high (1.550 mg L^{-1}), this being a consequence of possible water contamination with animal wastes from a livestock farm situated nearby [53].

The water quality of the Mures River in the Romania–Hungary cross-border area was evaluated during 2012 and 2013, and it has been found that nitrate levels are very high (7.3–27.3 mg L^{-1}), with the highest values being determined during autumn. Also, nitrite was found in high levels (0.12–0.3 mg L^{-1}). Hazardous nitrate and nitrite levels are due to inadequate management of nitrogen-containing fertilizers [54].

Another study [55] investigated the water quality of water from the Osam River, the second longest tributary of the Danube River in Bulgaria. The analyses were carried out, including four sampling points. The average values for ammonium nitrogen were: 0.136 mg L^{-1} (Osam River after Troyan), 0.125 mg L^{-1} (Osam River after Lovech), 0.187 mg L^{-1} (Osam River after Levski), and 0.166 mg L^{-1} (Osam River near Cherkovitsa). Average concentrations for nitrate nitrogen were: 0.685 mg L^{-1} (Osam River after Troyan), 1.043 mg L^{-1} (Osam River after Lovech), 2.311 mg L^{-1} (Osam River after Levski), and 2.534 mg L^{-1} (Osam River near Cherkovitsa). Nitrite nitrogen ranged between 0.011 and

0.037 mg L^{-1} in subjected sampling points. Total nitrogen concentrations were, as average: 0.987 mg L^{-1} (Osam River after Troyan), 1.582 mg L^{-1} (Osam River after Lovech), 3.150 mg L^{-1} (Osam River after Levski), and 3.356 mg L^{-1} (Osam River near Cherkovitsa).

Nitrogen pollutant species (nitrate, nitrite, and ammonium) were monitored during a period of three years in river systems (Aliakmon, Axios, Gallikos, Loudias, and Strymon) from Northern Greece. The results of the analyses indicated 0.21 mg L^{-1} (0.01–1.56 mg L^{-1}) nitrite–nitrogen, 0.38 mg L^{-1} (0.3–10.2 mg L^{-1}) nitrate–nitrogen and 1.22 L^{-1} (0.03–3.08 mg L^{-1}) ammonium-nitrogen, and soil leaching was considered as the main source of pollutants [56]. Other studies [57] developed in Greece aimed at monitoring water quality from the Nestos River basin. Among nitrogen species, nitrate level was the highest and ranged between 0.19 and 0.45 mg L^{-1}. Nitrite nitrogen and ammonium nitrogen were in the range 0.004–0.218 mg L^{-1} and 0.054–0.281 mg L^{-1}, respectively. Total nitrogen levels were between 0.83 and 1.75 mg L^{-1}. Based on these values, the water quality is moderate to good.

In France between 2000 and 2019, nitrate nitrogen levels in surface waters were stable and ranged between 2 and 5 mg L^{-1}. In the past, nitrate concentrations were higher, but after adopting proper agricultural practices, they registered a declining trend [58].

The quality of water from Miedwie Lake in Poland was evaluated the more so as it has tributaries, which are drainage ditches that collect water from agricultural areas. Nitrate concentrations range between 0.3 and 89.7 mg L^{-1} and ammonium 0.03–32.8 mg L^{-1}. Also, 93% of nitrogen loadings into lakes were attributed to agriculture [59].

Evaluation of nutrient levels from fourteen Italian lakes situated in Northern Italy was reported by Leoni and co-workers [60]. The determined average results were: 1.173 mg L^{-1} for total nitrogen, 0.593 mg L^{-1} for nitrate nitrogen, and 0.126 mg L^{-1} for ammonium nitrogen. These values were due to the use of fertilizers and intensive animal farming.

With respect to Finnish surface water, nitrate levels are low compared with other European countries, and since 1996, nitrate concentrations have been stable [49].

Over time, monitoring studies regarding nitrogen pollutant species for surface water have been carried out in all European countries, as it can be seen from the data presented above. The presence of nitrogen species in lakes and rivers is related mainly with animal farming or inadequate use of fertilizers, and declining of the concentrations occurs by adopting sustainable agricultural practices.

- *Actual situation in EU*

Nowadays, more than 30 years after the initiation of nitrates directive and despite legislation addressing nutrient pollution, nitrate water pollution is still an actual problem that needs a solution.

For instance, the average nitrate levels either in EU groundwaters (Figure 5) [17] or in rivers (Figure 6) [61] did not suffer a significant decrease during the monitoring period 2000–2021.

Contrariwise, ammonium nitrogen levels in EU rivers have decreased between 2000 and 2021 (Figure 7), according to data reported to the EU [62].

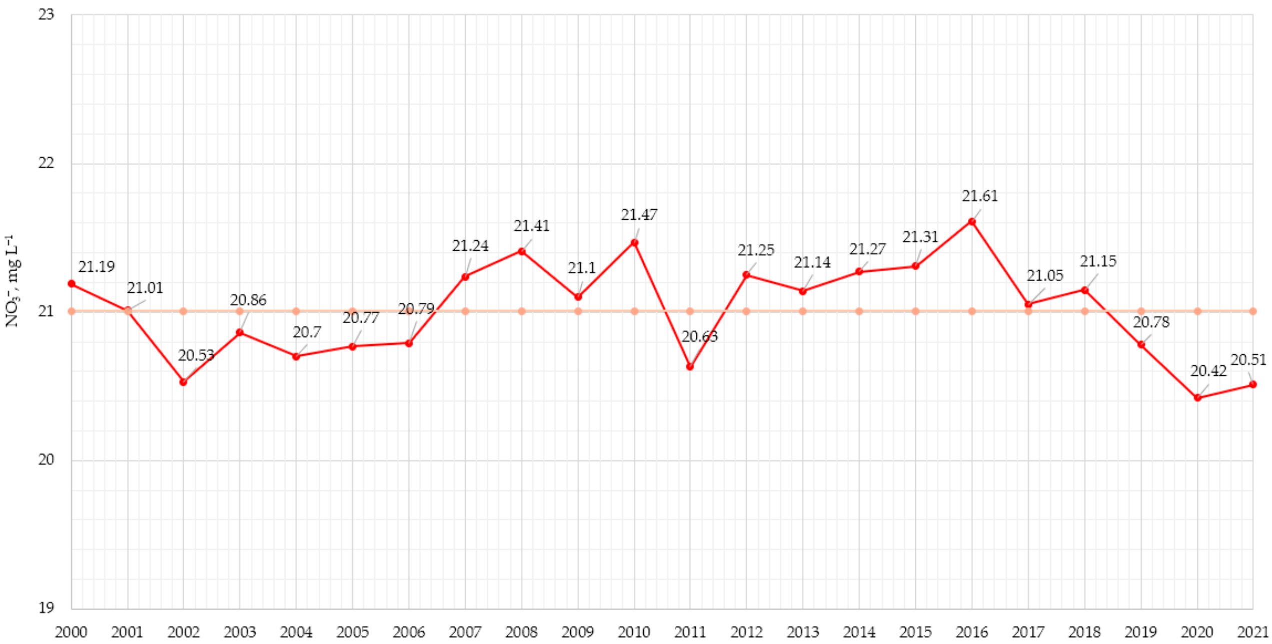

Figure 5. Nitrate levels in EU groundwater (1025 groundwater bodies). (orange line from graphic represents the average of the measurements during 2000 and 2021).

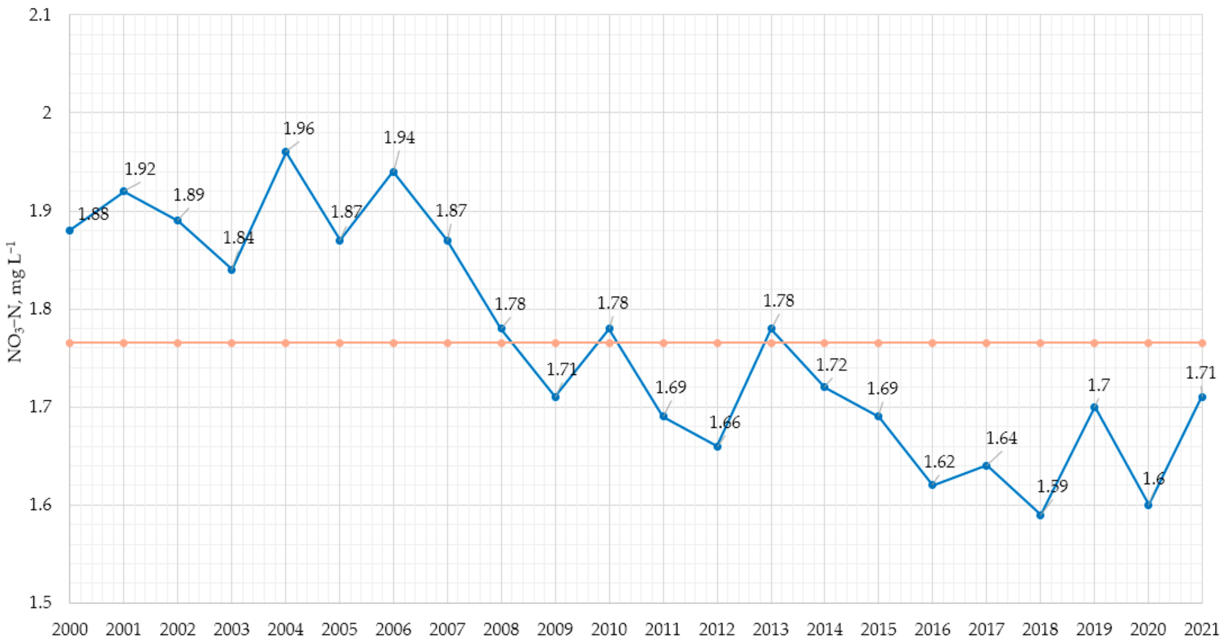

Figure 6. Nitrate nitrogen levels in EU rivers (1006 monitored sites). (orange line from graphic represents the average of the measurements during 2000 and 2021).

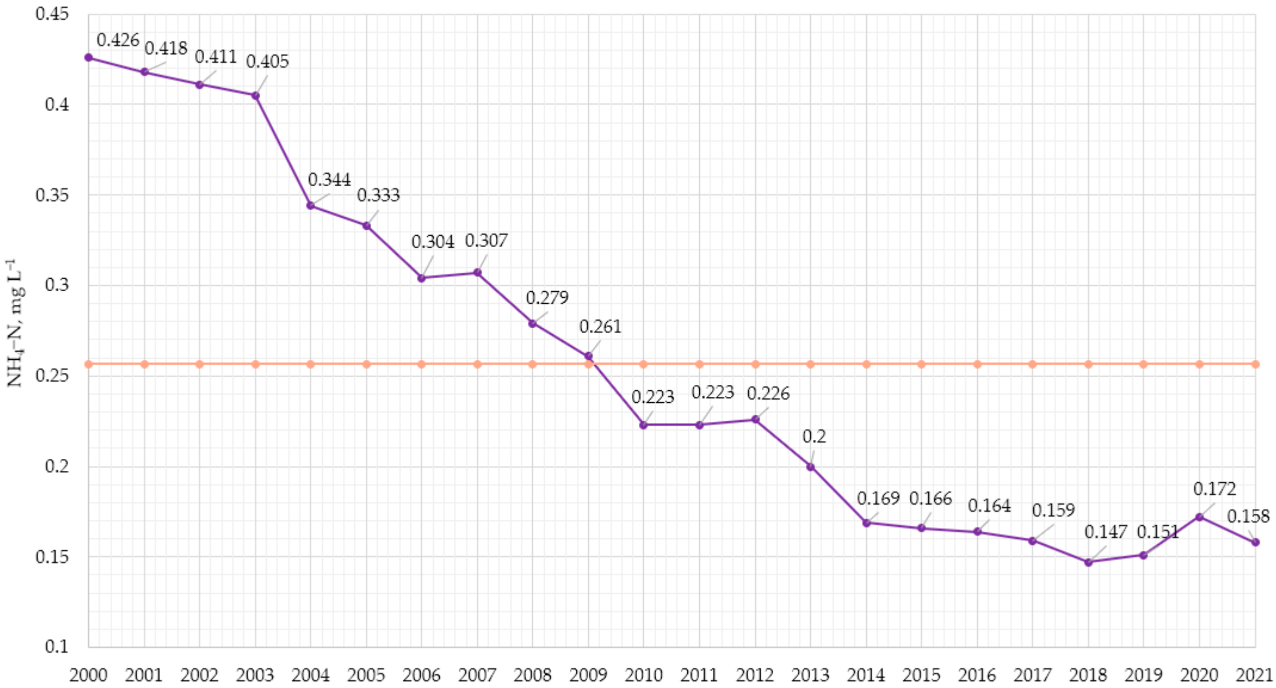

Figure 7. Ammonium nitrogen levels in EU rivers (773 monitored sites). (orange line from graphic represents the average of the measurements during 2000 and 2021).

2.3. Phosphorus Pollution

2.3.1. Phosphorus Forms and Route from Agriculture to Water Bodies

Phosphorus (P) is considered an essential macronutrient for plant growth, being involved in many processes, such as the synthesis of phospholipids and nucleotides, photosynthesis, respiration, redox processes, improving flower formation, the quality of fruits, and favouring increased resistance to plant diseases [63,64]. Consequently, in agricultural systems, limited availability of phosphorus in soil is very challenging for crop productivity. Anyway, addition of phosphorus beyond agronomic crop requirements has minimal effects on yield. Furthermore, according to Lizcano–Toledo and his team [63], more than 70% of the arable surface on Earth necessitates phosphorus supply to produce crops.

The presence of phosphorus in soil is related to minerals, commercial fertilizers, or organic manures, so both inorganic and organic forms are encountered in soil. Inorganic forms used by plants are either $H_2PO_4^-$ in acid soils or HPO_4^{2-} in alkaline soils. Phosphorus availability is maximum when soil pH is between 6 and 7. In addition, the level of this macronutrient available to plants is very low and varies from 0.001 mg L^{-1} to 1 mg L^{-1}. Regarding organic form, it varies from soil to soil, and it may represent 20 to 80% of the total phosphorus from the surface soil horizons [65].

Considering its immobile character in soil, phosphorus leaching downward in the soil profile occurs if its level is very high. This situation occurs when fertilizers and manure are applied excessively. Phosphorus enrichment of surface water is related to several factors such as soil type, high soil phosphorus, and soil hydrology [65].

Inorganic and organic forms of phosphorus from soil are transferred by runoff, erosion, and leaching to water and are encountered in dissolved (soluble form) or particulate forms (eroded soil particles) (Figure 8). Dissolved phosphorus is available to aquatic organisms and responsible for eutrophication. Particulate phosphorus is either unavailable or slowly available to aquatic organisms.

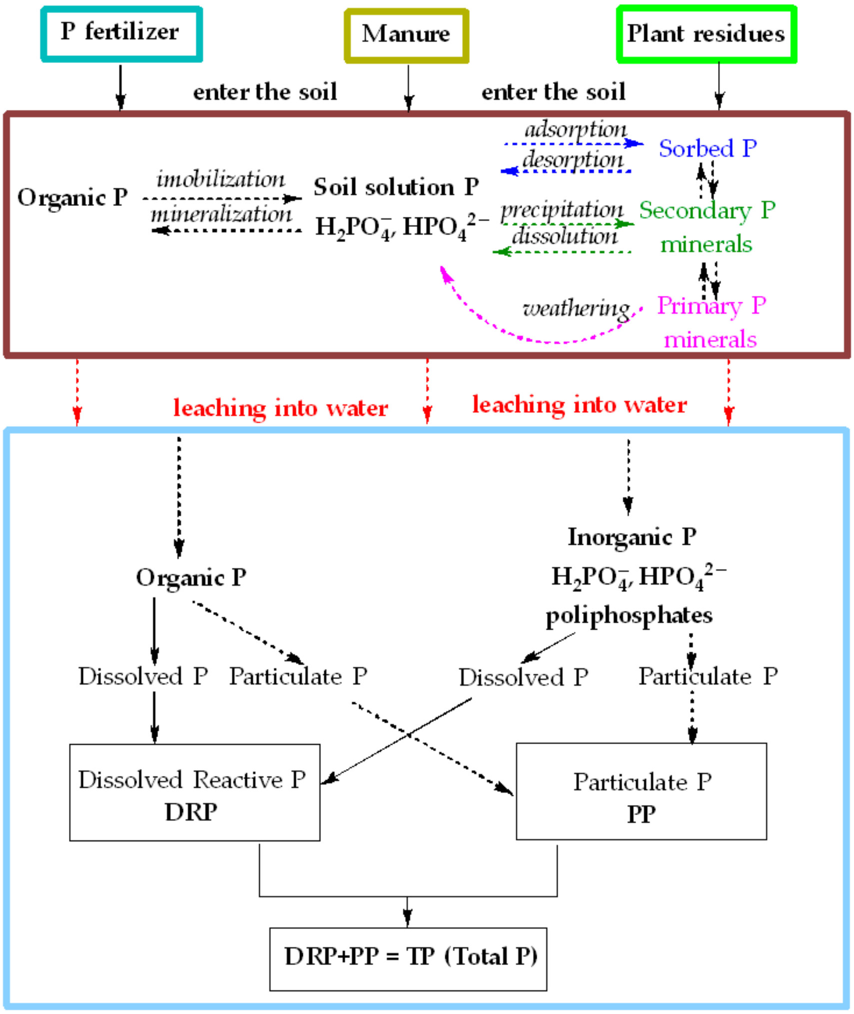

Figure 8. The forms of phosphorus in the soil and in the water.

A small quantity of dissolved phosphorus is found in soils naturally, while particulate form moves mainly by soil erosion. The best management tool suitable to ensure that soil does not accumulate large quantities of phosphorus is laboratory soil testing. The limits for soil phosphorus levels that are optimum for plant growth and those above which very high environmental risk is possible to appear are presented in Table 1 [65].

Table 1. Limits for soil phosphorus from agronomic and environmental perspectives (mg kg^{-1}).

Extraction Method	Optimum	Environmental
Mehlich-1	13–25	>55
Mehlich-3	25–50	>50
Bray-1	20–40	>75
Olsen	12	>50

2.3.2. Tracing Phosphorus Species in Water

The monitoring of phosphorus in the environment on the basis of stable isotopes is not an option since phosphorus has one stable isotope (^{31}P), two radioactive ^{32}P ($t_{1/2}$ = 14.36 days), and ^{33}P ($t_{1/2}$ = 25.34 days), and the rest to 22 are very unstable (with half-lives under one second) [66].

During time, there were some attempts to use radioactive isotopes, ^{32}P and ^{33}P, but there were drawbacks represented mainly by health risks. As phosphorus is usually bonded by oxygen, which has three stable isotopes, the scientists have identified a safe and efficient route to track phosphorus in aquatic environments and to understand better phosphorus cycling. This approach is based on stable oxygen isotope ratio $^{18}O/^{16}O$ within phosphate ($\delta^{18}O_{PO4}$), but there are many limitations.

Among the encountered difficulties is the lack of a library of reference samples with specific values and detailed information regarding isotopic sources from various geographical regions. Also, it is difficult to distinguish between $\delta^{18}O_{PO4}$ of chemical phosphorus fertilizers and phosphorus discharged from wastewater treatment plants [67]. Values for poultry, dairy, and swine manure were reported by Jacquemin and co-workers [68] as 18.5‰, 16.5‰, and 17.9‰, respectively.

Research developed by Granger and his team [69] provided information regarding phosphate sources in the River Taw catchment, where $\delta^{18}O_{PO4}$ varied between +17.1‰ and +18.8‰. Potential pollution phosphate sources were sampled and analyzed, and most had a narrow range of $\delta^{18}O_{PO4,}$ similar to those identified for river water. For example, dairy factory final effluent had $\delta^{18}O_{PO4}$ from +16.5 to +17.8‰, stored animal wastes +12.0 to 15.0‰, and inorganic fertilizers from +13.3‰ to 25.9‰.

Another study [70] investigated the phosphate origin from groundwater, and on the basis of $\delta^{18}O_{PO4}$ values, it was found that phosphorus leaching from orchard soil (intensive citrus cultivation) was the main enrichment source.

Concluding, in aquatic ecosystems, the use of $\delta^{18}O_{PO4}$ to discriminate among phosphate sources is still at an early stage and needs further studies to overcome all the limitations and ambiguities.

2.3.3. Phosphorus Levels Resulted from Agricultural Practices in Water Bodies from Europe

In natural waters, inorganic phosphorus is represented mainly by orthophosphate, PO_4^{3-}, which is accessible for organisms. Total phosphorus (TP) can be defined as the maximum potential concentration of bioavailable phosphorus [71], and its monitoring has great importance for aquatic studies and for characterization of trophic state for surface water (as it can be seen in Section 3. Effects of nutrient pollution on water and human health). In water bodies, inorganic phosphorus and TP are rarely higher than 0.1 mg L^{-1} and 0.5 mg L^{-1}, respectively [72].

One of the main contributors to phosphorus levels in surface water is the use of fertilizers for crops. For example, agriculture's contribution to phosphorus levels in European waters is 20–40% [73].

- ***Surface water***

Phosphate phosphorus levels determined in the Ebro River, Spain, during a long-term experiment were between 0.2 and 0.9 mg L^{-1}. It has been observed that phosphorus levels decreased over time due to the reduction of phosphate fertilizer application [50].

TP average levels reported for upper River Itchen, UK, ranged between 0.166 and 0.613 mg L^{-1}, the pollution sources being represented by agriculture, aquaculture, commercial growing of watercress, and WWTP effluent [74].

Lower levels for TP were determined for fourteen Italian lakes situated in Northern Italy and ranged between 0.003 and 0.144 mg L^{-1}, with an average of 0.022 mg L^{-1} [60].

The TP and orthophosphate contents in Głuszynka River in Poland during 2016 and 2018 were 0.08–1.76 mg L^{-1} and 0.05–0.62 mg L^{-1}, respectively. These values, combined with nitrogen species levels, led to the classification of the ecological status of the waters from this river as "poor" [75].

In the case of two rivers from Romania, Timis and Bega, the TP average concentrations were 0.05–0.171 mg L^{-1} and 0.099–0.418 mg L^{-1}. Among pollution sources for these two rivers is mentioned agriculture [52]. Phosphate phosphorus level from Mures River in Romania Hungary cross border area was very high, the highest concentration being 1.2 mg L^{-1} [54].

Phosphate phosphorus and TP determined for Osam River, Bulgaria, were in the range (as average) 0.031–0.075 mg L^{-1} and 0.039–0.091 mg L^{-1}, respectively [55].

Monitorization of TP during 1982 and 2022 in the case of rivers from Germany evidenced a decrease by a round one-third, and even though extreme levels of TP are rare, to meet the requirements imposed by WFD, optimization of phosphorus management in agriculture is needed [76].

A study [56] conducted during 1997 and 2000 in Northern Greece that included several river systems (Aliakmon, Axios, Gallikos, Loudias, and Strymon) evidenced TP 0.57 mg L^{-1} (as average) and phosphates 0.22 mg L^{-1} (as average).

In another study [57] developed also in Greece, the phosphate phosphorus and TP contents for water from the Nestos River basin ranged between 0.008 and 0.019 mg L^{-1} and 0.037 and 0.132 mg L^{-1}, respectively. The water quality from this perspective is high to good.

Phosphate phosphorus in surface waters in France has decreased from 2000 to 2019 by 37%, as a consequence of reducing phosphate fertilizer use and also due to the ban of phosphates in laundry detergents [58].

Considering phosphorus levels in the Tisza River from Hungary, it has been observed during time a wide range of values. For example, in 1995, TP was 0.067 mg L^{-1} and phosphate phosphorus was 0.045 mg L^{-1}. Later, in 2001, the values increased, and TP was 0.128 mg L^{-1} and phosphate phosphorus was 0.050 mg L^{-1}. During 2001 and 2011, the phosphorus levels decreased, and in 2012, TP was 0.220 mg L^{-1} and phosphate phosphorus was 0.0373 mg L^{-1}. In 2023, TP was 0.073 mg L^{-1} and phosphate phosphorus was 0.040 mg L^{-1} [77].

Excess of phosphorus in surface waters is a global problem since eutrophication is one of the main effects, with severe consequences such as acidification, hypoxia, and the development of harmful algae blooms. According to the presented data, phosphorus levels vary a lot between countries and during time. Its decreasing trend is a consequence of optimization of phosphorus fertilizer doses or banning of phosphate use.

- ***Actual situation in EU***

Considering TP reported levels in EU lakes (Figure 9) and the soluble phosphates in EU rivers (Figure 10) between 2000 and 2021, it may be observed a decreasing trend starting with 2011 approximately, when was banned the addition of phosphates in detergent formulation. However, the presented results suggest that in lakes phosphorus level is lower than in rivers, the more so as reported values for lakes are TP form, which include soluble phosphates. Furthermore, another report [11] indicates also that phosphorus levels in EU rivers are higher than in EU lakes.

The major source of phosphorus in EU lakes is still diffuse runoff from agricultural land, followed by phosphorus stored in sediments [78]. In addition, the load of phosphorus in water is associated with human activities, and it is considered that without them, the concentrations would be 5–10% from the actual levels [73].

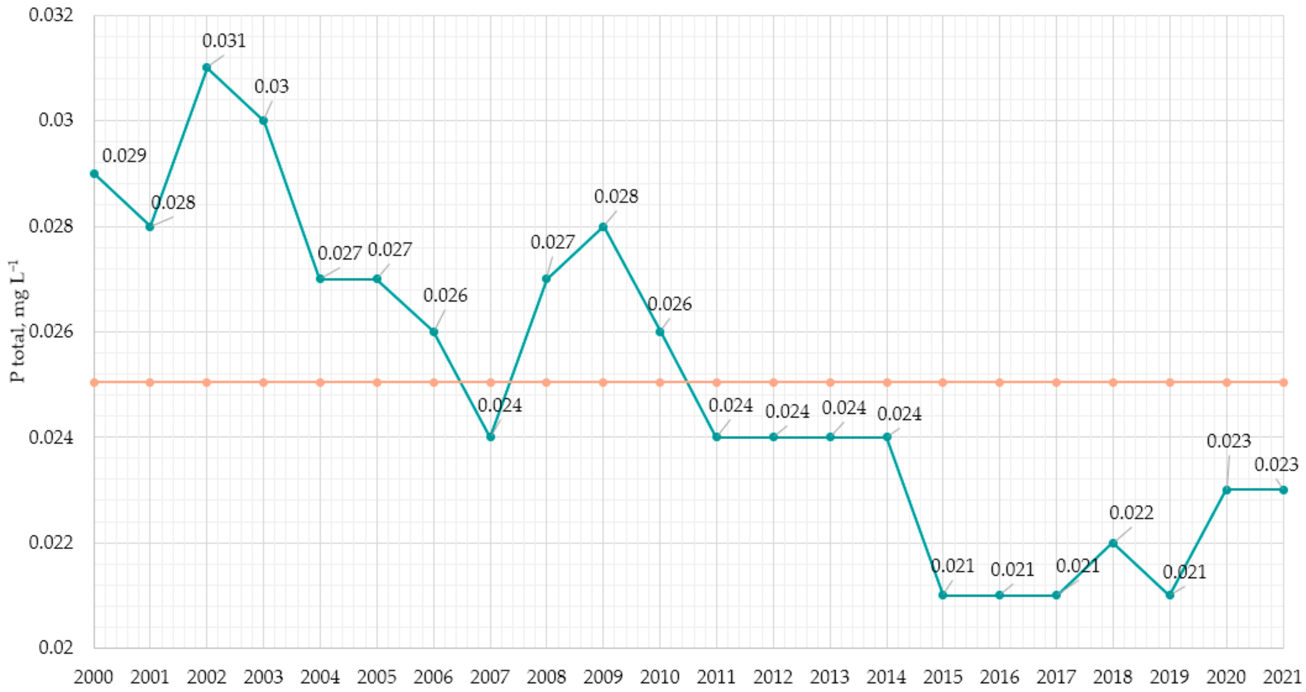

Figure 9. Phosphorus (total form) levels in EU lakes (341 monitored sites). (orange line from graphic represents the average of the measurements during 2000 and 2021).

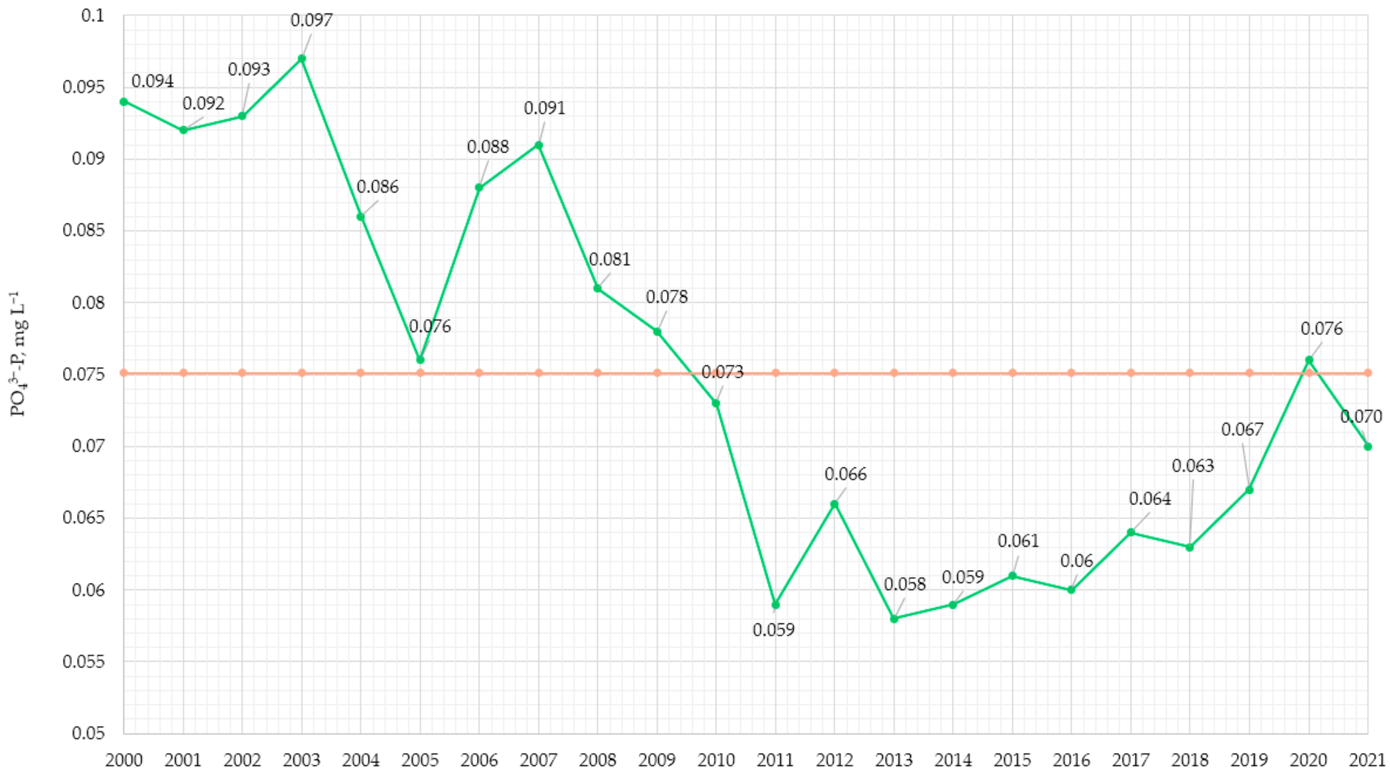

Figure 10. Phosphate phosphorus levels in EU rivers (693 monitored sites). (orange line from graphic represents the average of the measurements during 2000 and 2021).

3. Effects of Nutrient Pollution on Water and Human Health

3.1. Eutrophication

Excessive amounts of nitrogen and phosphorus resulted from nitrogen and phosphorus-based inputs can be washed from farm fields and enter easily into waterways after rain or after snow melting. They also may leach through the soil and find groundwater in time. Consequently, the presence of these nutrients in water courses stimulates excessive algal growth; this phenomenon is known as eutrophication.

According to the Nitrates Directive [8], eutrophication "means the enrichment of water by nitrogen compounds, causing an accelerated growth of algae and higher forms of plant life to produce an undesirable disturbance to the balance of organisms present in the water and to the quality of the water concerned".

The trophic state for surface waters is set on the basis of nutrients' concentrations, namely total nitrogen and total phosphorus, and chlorophyll "a" concentration. Nürnberg's classification and trophic limits set by him [79] were used during time, but nowadays these limits are set by legislation [12] (Table 2).

Table 2. Trophic state for surface water according to nutrients and chlorophyll "a" level.

Trophic State	Total Phosphorus, mg L^{-1}	Total Nitrogen, mg L^{-1}	Chlorophyll "a", μg L^{-1}
ultraoligotrophic	<0.005	<0.2	<1
oligotrophic	0.005–0.01	0.2–0.4	1–2.5
mesotrophic	0.01–0.03	0.4–0.65	2.5–8
eutrophic	0.03–0.1	0.65–1.5	8–25
hypertrophic	>0.1	>1.5	>25

There are many studies regarding the level of phosphorus in water, but there is no agreement among researchers about the threshold value that generates eutrophication. Some studies indicate that 0.01 to 0.03 mg L^{-1} of phosphorus can enhance the development of harmful algae [80]. This situation is far from being solved since eutrophication is generated by a combination of factors, and algal blooms depend strongly on the environmental conditions in the studied area.

For example, for water from Moara Domneasca pond located near agricultural areas in Ilfov, Romania, were reported 2.445 mg L^{-1} phosphate–phosphorus, 0.114 mg L^{-1} nitrite–nitrogen, 0.386 mg L^{-1} nitrate–nitrogen, 7.986 mg L^{-1} ammonium–nitrogen, and algal blooms were evidenced at the moment of water sampling [81].

Many studies investigated which nutritive element, nitrogen or phosphorus, must be controlled to prevent eutrophication. Long-term studies indicated that algal bloom and eutrophication are strongly influenced by reducing phosphorus, and there is no evidence that controlling nitrogen inputs would assure a better management of eutrophication [82]. Therefore, phosphorus is one of the primary limiting factors of eutrophication.

A novel approach to understanding eutrophication in aquatic ecosystems is based on the oxygen isotope composition of phosphate ($\delta^{18}O_{PO4}$) combined with isotopic analysis of oxygen and nitrogen from nitrate ($\delta^{15}N_{NO3}$, $\delta^{18}O_{NO3}$) and nitrogen isotope from ammonium ($\delta^{15}N_{NH4}$) [83].

Nitrogen and phosphorus enrichment encourage the growth of algae and different aquatic plants, leading to hypoxia, oxygen depletion, fish mortality, and poor water quality. Algal blooms limit penetration of sunlight into deep water layers; the photosynthesis does not occur, and finally aquatic plants die [84]. A representation of processes that occur during eutrophication is presented in Figure 11.

Blue–green algae (cyanobacteria) are a group of photosynthetic prokaryotes that appear in late summer and early fall on top of ponds or other stagnant waters [80]. Their presence is unwanted since many of them have the capacity to produce toxins dangerous to humans and animals [85,86].

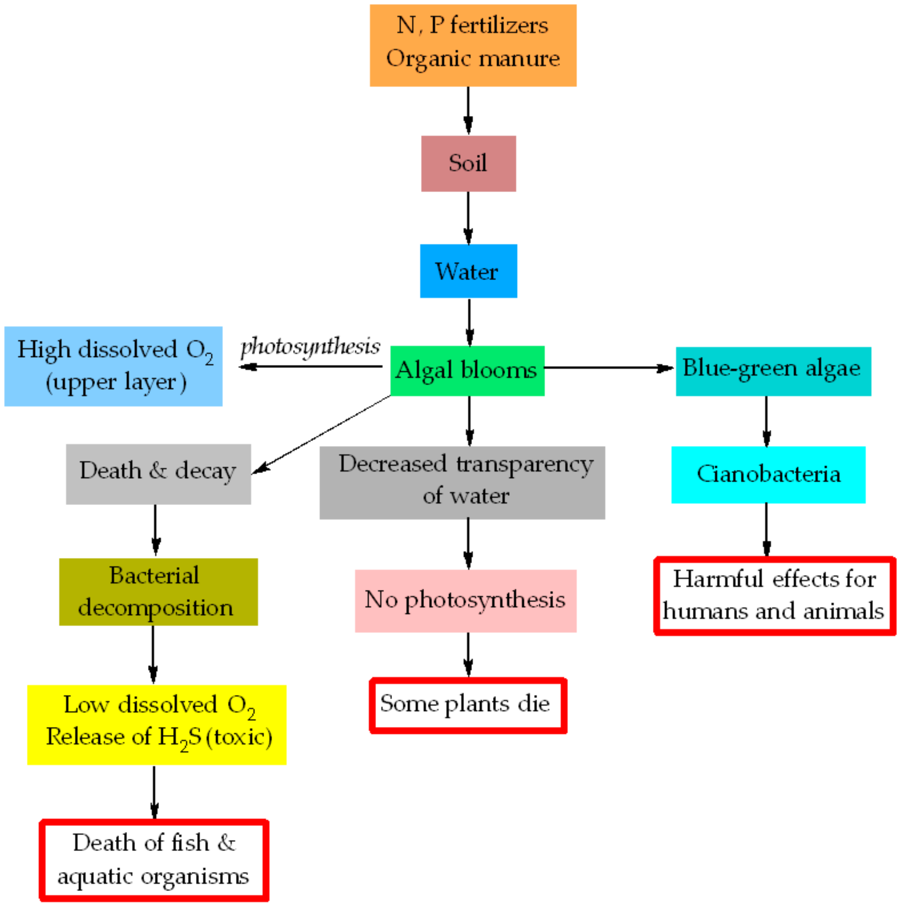

Figure 11. Eutrophication—causes and effects.

Beside these, dense algal growth presents economic impact because their presence can limit access to waterways. Unpleasant odors that may appear doubled by the toxicity of green–blue algae affect recreational activities (fishing, swimming, and boating).

3.2. Adverse Effects on Human Health

Consumption of drinking water with elevated nitrate levels may trigger a plethora of diseases and manifestations, as it is presented below.

- Methemoglobinemia (blue baby syndrome) has been described firstly by Comly in 1945 [87]. This manifestation occurs mainly to infants and the elderly and consists in the reduction of nitrate to nitrite, followed by the oxidation of ferrous ion (Fe^{+2}) from hemoglobin (Hb) to ferric ion (Fe^{+3}) in methemoglobin (MetHb), which is unable to bind oxygen.

$$\underset{\text{nitrate}}{NO_3^-} \xrightarrow[\text{acidic pH}]{\text{bacterial reduction}} \underset{\text{nitrite}}{NO_2^-} + \underset{\text{hemoglobin}}{Fe^{+2}Hb} \rightarrow \underset{\text{methemoglobin}}{Fe^{+3}MetHb}$$

Methemoglobin represents 1% of the total hemoglobin in the case of a healthy adult, and it could be slightly higher in the case of newborn infants. When methemoglobin levels in infants are around 3%, there is obvious cyanosis [88]. Other signs and symptoms of methemoglobinemia are headache, dizziness, fatigue (MetHb = 0–45%), coma, convulsions (MetHb = 45–55%), and high risk of mortality (MetHb > 60%) [89].

In order to safeguard human health against the harmful impacts of nitrate in drinking water, the World Health Organization (WHO) specified that the permissible limit is 50 mg L^{-1} [90], and the US Environmental Protection Agency recommends a value of 44 mg L^{-1} [30].

Some specifications regarding nitrate levels in water [34] are presented in Figure 12.

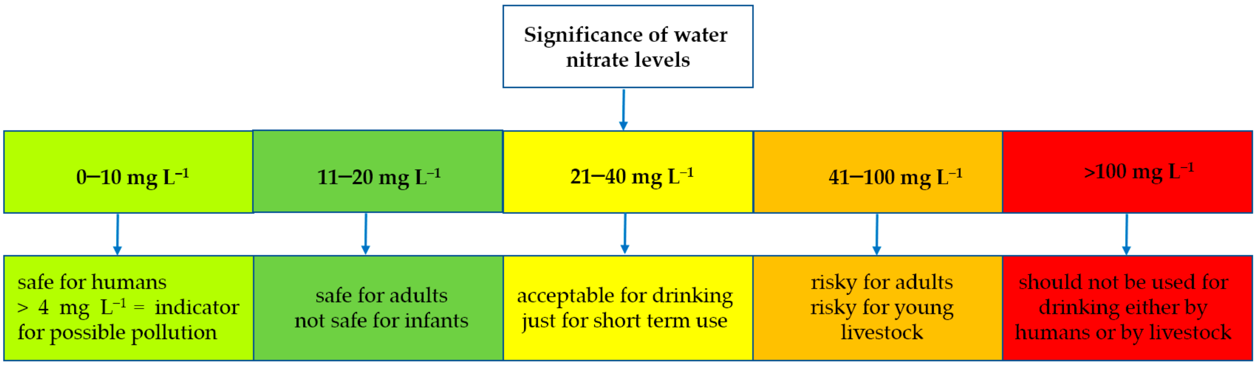

Figure 12. Nitrate levels and significance for health.

- Formation of potential carcinogenic N-nitroso compounds

Under the influence of the microflora and the acidic medium in the stomach, nitrates, are converted into nitrites which further transform into *N*-nitroso compounds (*N*-nitrosoamine and *N*-nitrosoamide) with carcinogenic potential [91]. Consequently, there are studies that relate the consumption of nitrate-contaminated drinking water with various types of cancer [92–95].

- Other health effects

Literature studies present data related to nitrate ingestion from drinking water and the prevalence of thyroid disfunction, indicating an increased prevalence of hypothyroidism but not hyperthyroidism [96,97]. Also, there are studies that report a link between nitrate exposure and adverse reproductive outcomes (congenital abnormalities, low birth weight, birth defects) [98], lowering blood pressure [99].

4. Strategies for Managing Agricultural Practices to Minimize Water Pollution

Considering the effects of agricultural pollution, especially with nutritive species, on groundwater, rivers, or lakes, it is necessary for farmers to adopt some strategies by which the effects of fertilizer use must not affect the quality of the environment.

Some of the presented approaches are relatively easy to implement, offering savings due to the use of smaller amounts of inputs, while others require higher expenses, lower yields with lower quality in some cases, and implicitly lower profit.

An efficient strategy for preventing or reducing the pollution resulted from agriculture will address both the pollution sources and the pathways by which pollutants enter watercourses. Lately, organic farming is considered a sustainable measure for reducing pollution resulted from agriculture [100,101].

A brief presentation of the most common methods adopted by farmers is presented in Figure 13.

Accurately calculating crop nutrient requirements improves crop yields, reduces costs to the producer, and minimizes the risk of water pollution. To fulfill this objective online, there are many crop calculators that can be accessed free of charge [102,103].

In order to prevent water pollution by the use of **phosphorus-containing fertilizers**, there are some approaches, as follows: (i) the proper amount and timing of phosphorus inputs; (ii) type of used fertilizer; (iii) use of phosphate inputs with less water-soluble phosphorus [104]. For example, the use of monoammonium phosphate (MAP), diammonium phosphate (DAP), simple superphosphate (SSP), and triple superphosphate (TSP) poses environmental impact due to their high solubility. Contrariwise, magnesium ammonium phosphate (struvite) has lower solubility, and its application is safe from an environmental perspective [63]. A slow and very slow-acting such as basic slag, a low-phosphate fertilizer with a liming effect, and use to improve soil nutrient levels over an extended period, especially as part of a long-term soil improvement strategy.

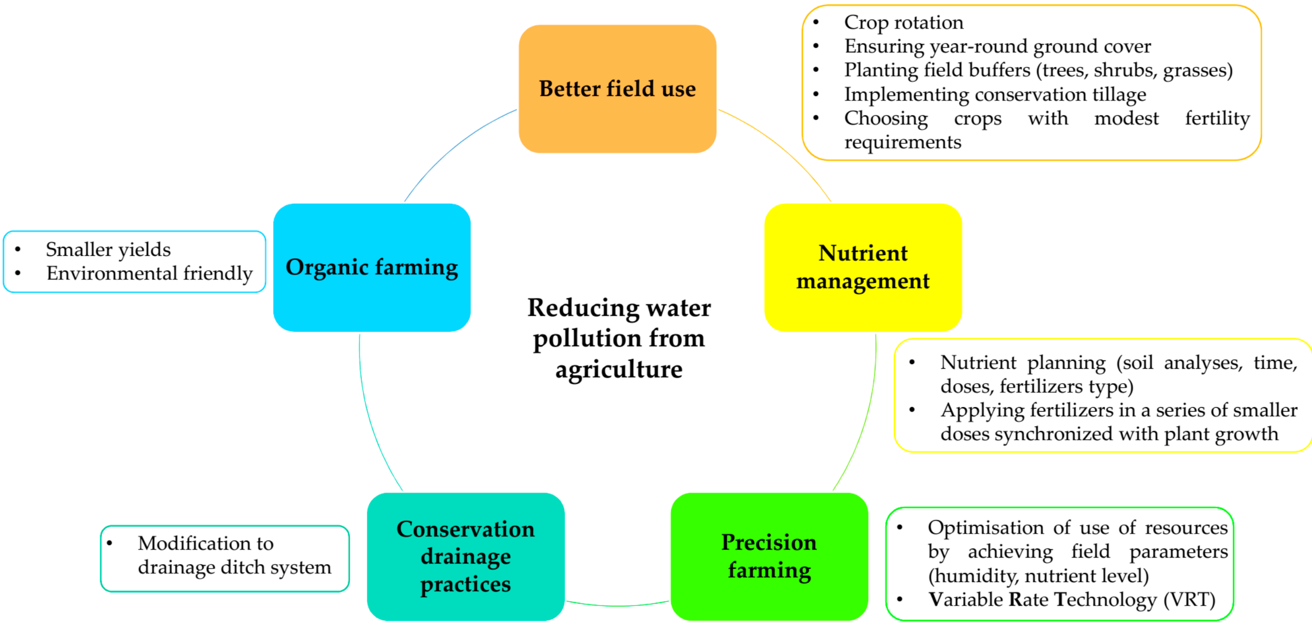

Figure 13. Strategies for reducing water pollution from agriculture.

Other recommendations [104] to limit the transfer of phosphorus to water sources consist in:

- Identification of the critical level of plant-available phosphorus for each farming system and soil;
- Minimizing the transport of phosphorus to water by minimizing soil erosion;
- Before application of inputs, the farmers should consider existing soil phosphorus level combined with contributions from organic manures, sewage sludge, and chemical fertilizers;
- Plant-available phosphorus should not increase much above critical value.

Mitigation of the environmental impacts of nitrogen use in agriculture is also necessary since the levels of inorganic nitrogen species (nitrate, nitrite, and ammonium) in water bodies are still high even if there are regulations that should protect water quality.

Some **nitrogen management strategies** that would abate water nitrogen pollution are:

- 4R nutrient stewardship (right fertilizer type, right amount, right placement, and right time) [105];
- Soil testing before application of nitrogen fertilizers to apply only the amount the crop needs;
- Application of nitrogen fertilizers under proper environmental conditions to prevent runoff;
- Use of environmentally friendly fertilizers that delay the release of nitrogen into soil [106];
- Application method (incorporation of the fertilizers into the soil or injecting).

Also, for optimization of fertilization strategies, effective guidance is assured by different models on nitrogen uptake and leaching [107]. Excessive use of nitrogen inputs is the main factor in high nitrate levels in groundwater, and some of the models are capable of predicting nitrate leaching from fertilizer treatments. For example, the Hydrus-2D/3D model is able to simulate the impact of current management practices on nitrate leaching in orchards. It predicted that nitrate leaching would decrease by 37% when drip irrigation is used instead of a sprinkler [108]. Model Hydrus-1D simulated that a 50% reduction in added nitrogen inputs would reduce nitrate in groundwater by 70% [109].

In addition, proper storage and handling of mineral and organic fertilizers will reduce the risk of causing water pollution resulting from fertilizer spills and container failures.

Manure storage need special attention considering that liquid and slurry manure can easily drain and produce nutrient losses into the environment. If not managed properly, nutrients from manure influence negatively the environment (soil, water, and air). For instance, 80% of the total load of nitrogen in surface water is attributed to agriculture. Also, gaseous products (ammonia and nitrous oxide) are lost in the air from manure during storage and land application [110]. Furthermore, ammonia volatilization near animal farms may become a water pollutant when it returns dissolved by rainfall.

Considering these aspects, livestock farmers must respect a nutrient management plan and comply with manure application limits of 170 kg ha^{-1} year^{-1} (with some exemptions if manure disposal does not affect ecosystems) and obey additional regulations related to application timing, method, soil type, climatic conditions, rainfalls, and irrigation [6,8,111]. Among strategies to fulfill this objective, it is worth mentioning management of runoff, leaching from stockpiled manure and from storage pits, and using a clean-water diversion system. In addition, there are reported methods (anaerobic digestion of manure and subsequent digestate processing technologies) that stabilize manure in order to be applied in a safe manner to soil [112].

Concluding, preventing water pollution with nutrients resulted from agriculture is challenging because nutrient management is cost-effective, the more so as the use of environmentally friendly fertilizers implies higher costs. Adopting certain agricultural practices and strategies will decrease pollution levels but the crop yield as well.

5. Conclusions and Further Perspectives

Nowadays, agricultural practices require greater consideration due to their important contribution to water pollution, the more so as it seems that they seriously impact surface and groundwater quality with consequences on human health.

The regulations surrounding water pollution and the strategies farmers can employ are supposed to reduce pollution, but despite regulatory actions, such as the nitrate directive, there is still little progress on this issue. The levels of nutrient species encountered in European waters (groundwater, lakes, and rivers) did not decrease significantly. Notwithstanding the sustained efforts made by each country and specialists involved in this action during 20 years, nowadays 40% of the European water bodies do not meet the requirements imposed by WFD [55].

Adoption of good agricultural practices is difficult to implement because it would be relatively costly for farmers to adopt (cover crops, reduced tillage, and land retirement). Still, farmers need to be trained to adopt some measures to prevent pollution, even though this would possibly affect the crop yield and quality. Decreasing fertilizer rates is considered quite efficient considering that the relationship between fertilizer doses and profit is flat near optimum [113]. Even though manure is a great source of nutrients for crops, defective management of manure can degrade the environment. Consequently, according to European legislation, farmers have the responsibility to manage manure storage and application in order to prevent water pollution.

Despite monitoring programs and legal policies, nutrient pollution from agriculture is still an actual problem, difficult to solve totally, considering the importance of nutrients for crops and yields and the increased demand for food. Therefore, it is necessary that mitigation of nutrient pollution from croplands continue, and efforts should be focused on promotion of safe farming practices, improvement of nutrient management techniques, and continuous monitoring of water quality.

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